A SPECTROSCOPIC INVESTIGATION OF MOVING STRIATIONS IN AN ARGON GLOW DISCHARGE

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Lieutenant, United States Navy

Submitted in partial fulfillment of
the requirements for the degree of

MASTER OF SCIENCE
IN
PHYSICS

United States Naval Postgraduate School
Monterey, California

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ABSTRACT

An investigation of moving-striations in an argon glow discharge using a Baird-Atomic Plasma Spectrograph, was conducted to determine the time differences for appearance of spectral lines with different excitation potential. The procedure utilized photographs of oscilloscope traces comparing the 8115A line of argon with the 7372A and 4158A lines. These photographs were analyzed to determine the time differences within the striations at the points of maximum and minimum intensity. For discharges in a tube of 26.5mm diameter, with gas pressures in the range of 2mm to 7mmHg pressure, the results indicate the line of lower excitation energy (8115A) leads the higher excitation lines (7372A and 4158A) by 22 to 30 microseconds at minimum intensity and lags the higher excitation lines by 27 to 35 microseconds at the maximum intensity. It was also noted that the time of intensity rise for the striation was dependent upon the pressure of the gas discharge.

TABLE OF CONTENTS

LIST OF ILLUSTRATIONS

1. Introduction

1.1 Gaseous Discharges and Striations

Gaseous discharges and their properties, once a scientific curiosity, have assumed extreme importance in recent years, especially in the field of space research and in the development of a controlled fusion reaction as a source of energy for future generations. Although gaseous discharges and some of the phenomena associated with them have been observed for many years, no satisfactory theory has been developed to explain their existence. The interactions between atomic particles and their associated fields are extremely complex, and even with the strides made in the last two decades, there are still many unanswered questions.

On applying an electric field between two electrodes in a tube containing a gas, a very small current exists until the potential applied reaches some minimum value called the sparking potential. At this point the current increases many-fold and the system is characterized by the appearance of emission of light from the tube. Once the emission has started, the potential required to sustain the discharge is decreased. Because of the appearance of the discharge, the operating conditions of small current and moderate voltage have been given the name glow discharge region.

Now if we limit the gas in the tube to low pressure in the vicinity of a few mm of Hg, we can depict the appearance of the discharge into regions of low and high light intensities. Proceeding from the cathode out to a few cm, we have a primary dark space (Aston Dark Space), followed by the cathode glow which fades into

the cathode dark space (Crookes Dark Space); this is followed by the negative glow which fades into the Faraday Dark Space; then for a distance of several cm there is a long glowing region, where the electric field is small, called the positive column; and in the final few cm in front of the anode there is the anode glow and dark space.

Common to many gases, especially the inert or rare gases, is the appearance in the positive column of alternate bands of lightness and darkness called striations. These can be either moving striations, stationary striations, or a combination of both. The phenomenon of moving striations is that segment of the study of gaseous discharges with which this paper will be concerned.

Significant experimentation has been conducted in the field of moving striations since about 1925. A good resume of these may be found in Cooper's thesis. [1] Because the striations depend on the geometry of the tube, the material of the electrodes, and the purity of the gas, there was a certain amount of seemingly conflicting results in the early works. Consequently, it has not been until the last 20 years that important strides have been made in the study of striations. To mention but a few: Cooper and Oleson [2] in their study of the boundary conditions of the striated regions; Donahue and Dieke [3,4] on the modes of striations, negative striations, and phase differences of spectral line intensities; Pekarek [11,12] with his artificially induced "waves of stratification"; Robertson [13] who has developed the most substantial theory on moving striations, and who in experimentation with Hakeem [14] has demonstrated that metastable states must be present in

order to have sustained striations. Many others, too numerous to mention here, have conducted experiments on striations under various conditions using spectrographic techniques, Langmuir probes, rotating mirrors, and photomultipliers.

1.2 Phase Differences of the Spectral Lines in the Striated Region

Donahue and Dieke [3] observed in a mercury discharge, that the maximum intensity of the 2537A line of low excitation potential leads the 4047, 4358, and 5461A lines of higher excitation potential by about 15 microsec. They explain this time difference by a qualitative discussion of how ionization occurs in the positive column and how the striations propagate in the tube from anode to cathode. Briefly stated their explanation proceeds as follows: Let us assume that the only levels excited to any appreciable extent in the positive column by electron collision are the lower energy levels, some of which are metastable, and that the major process by which atoms are excited to a higher level or to ionization is by further electron collision with these atoms in metastable states. Now suppose that at some point in the positive column there exists a positive space charge peak where the population of metastable atoms is low. Furthermore, there is a concentration of atoms in metastable states at a small distance toward the cathode from this peak; and closer to the cathode there is a maximum rate of production of metastable states by electron collision. Now if we observe the travel of the electrons in the positive column moving from cathode to anode, they will approach the vicinity of a positive space charge caused by ionized argon atoms. These electrons have sufficient energy that, in colliding with the atoms in the leading

part of the positive space charge, they will excite these atoms to the lower and metastable levels. The electrons would, consequently, suffer a drop in energy and little excitation of the atoms would occur in the next part of their travel. Because of the concentration of ions ahead of them, they would again increase in energy. As a consequence of the collisions of the electrons proceeding them, the electrons would find themselves in a concentration of metastable atoms. Collision with these atoms would cause some atoms to be ionized and others to be excited to higher energy levels. The electrons would then move into a region of highly excited and ionized atoms. No further ionization of significance would occur due to the low concentration of metastable states and the low energy of the electrons. Finally the electrons loose their energy in the negative field on the anode side of the space charge; the ions either recombine or diffuse to the walls of the tube. This, of course, is a continuous process with the space charge configuration propagating toward the cathode.

The consequence of the above argument is that one would expect the maximum intensity for the lower energy levels to occur earlier in time, or to the cathode side of the striation, before the maximum of intensity for the higher energy levels.

Experiments to confirm this in the case of argon were conducted at the U. S. Naval Postgraduate School from 1956-58 by Pigg and Burton [9,10], McDonnell and Sherman [8], and Kolkhurst and Strong [7] using a Gaertner Model 1231 spectrometer equipped with a 1P21 RCA photomultiplier. The results of these experiments were not conclusive, and there was disagreement among the three groups.

It is due to these inconclusive results that the present investigation was conducted.

2. Experimental Techniques and Procedures

A Baird-Atomic Plasma Spectrograph was used to analyze the various spectral lines. The instrument has a range of 1280A to 22630A with an interval of approximately 2800A per grating setting. The discharge tube was oriented perpendicular to the spectrograph's optical axis, and the light emitted from the discharge tube was focused on the spectrograph entrance by means of a lens mounted on an optical bench. The light incident on the entrance was restricted by an adjustable vertical slit placed in front of the discharge tube. (See Fig. 3)

In selecting the transition levels to be observed, it was necessary to consider the limited range of the Baird-Atomic Spectrograph (2800 Angstroms)at one grating setting, the proximity of other lines in order to prevent the irradiation of the phototube by more than the specified line, the actual physical space available in the spectrograph for mounting the phototubes in close proximity of each other, and of primary importance, the excitation potential of the upper level of transition. The plates previously photographed by Kinley and O'Malia [6] in spectral work using the Baird-Atomic Spectrograph on argon discharges, as well as the tables they prepared, were used in selecting the following spectral lines:

Wavelength	Transition	Excitation Energy
7372.119A	$4d_{34}$-$4p_{23}$	14.78ev
4158.591A	$5p_{12}$-$4s_{12}$	14.47ev
8115.311A	$4p_{23}$-$4s_{12}$	13.02ev

(Details of the experimental apparatus are given in Appendix III)

Photomultiplier tubes mounted in the spectrograph were used to observe the spectral lines; an RCA 7102 photomultiplier tube for the 8115 and 7372A lines and an RCA 1P21 for the 4158A line. The 1P21 tube had a considerable higher noise level than the 7102 tube. This was not, however, a limiting factor.

The signal was maximized by adjusting the photomultipliers, aligning the slit and lens, and manually making small adjustments of the grating. Once the signal was maximized, the discharge current was adjusted to obtain stable striations. In general, the lowest possible current was used because the striations were more stable at lower currents. At low gas pressure, it was extremely difficult to obtain completely stable striations. Even though they appeared stable on the rotating mirror, they fluctuated very slightly back and forth on the scope and spread out, presenting a broad signal when observed with continuous sweep operation. The fluctuations and spreading decreased at higher pressures.

By masking the slits in front of the tube, it was noted that approximately 5mm of the discharge diameter was producing the signal on the oscilloscope, although the discharge filled about two-thirds of the 25.6mm discharge tube.

The spectral lines were observed in pairs and simultaneously presented on the dual trace type 551 Tektronix oscilloscope. The top trace (8115 line), inverted for ease in scanning of the photographs, was used to trigger the signal. (See Fig. 1) The horizontal

scale was set at 0.5 millisec/cm* and the vertical scale was adjusted according to the signal strength of the phototube output. The high-frequency filter cutoff was set at 20-60 kilocycles. Once stable striations were obtained, the scope was placed on single sweep and a Polaroid camera (time exposure, f-8 lens setting) with type 47 (ASA4000) film was used to photograph the single sweep pictures.

From a total of approximately 500 photographs, 180 were selected for scanning with a McPherson Instrument comparator. There were 15 pictures scanned for each operating condition observed, from which about 45 to 60 measurements of phase shifts at both maximum and minimum intensity could be obtained. (A sample of a picture used for scanning is shown in the bottom half of Fig. 1) The data were further refined by rescanning and eliminating those points where there was an appreciable noise level. For a given set of pictures, this meant that from 5 to 50 percent of the points were discarded.

*In order to facilitate scanning procedures and to obtain the maximum amount of information from one picture, the 0.5 millisec/cm scale was used. At the lower scales, it was more difficult to resolve time differences due to the decreased slopes of the signal at the maximum and minimum intensities.

3. Analysis of Data

Throughout the observed pressure range (2mmHg-7mmHg), the results indicate that there is a definite phase difference between the lines of different excitation levels, both at the point where the striation light intensity commences its rise and at the point of maximum intensity. There exists a 23 to 30 microsec time difference (the lower excitation line leading) where the striation starts to rise, and a 27 to 35 microsec time difference (the higher excitation line leading) at the point of maximum intensity. In other words, the lower levels are excited first and reach their maximum excitation later in time than the higher levels. Fig. 1 displays a typical photograph illustrating the above. Appendix I lists the average values of phase difference for the 8115 and 7372 lines for various pressures. The phase differences for the 4158 and 8115 lines show this same trend. The values in Appendix I are based on an average of 41 measurements for the phase differences at minimum intensity and an average of 27 measurements for the phase differences at maximum intensity. It was originally expected that the measurements would assume a normal distribution; however, even after the data had been refined by eliminating points of appreciable noise level, the distribution of values was contrary to expectations. There appears to exist a randomness of values inherent in the physical system other than that of a statistical nature. An additional reason for the large variation at measurements for maximum intensity is the difficulty in deciding where the signal maximum occurs. The slope is more gradual than the sharp slope at minimum intensity so that

it was more difficult to measure the point of peak intensity.

Attempts to correlate the phase difference as functions of pressure and current were inconclusive.

The period of the striations was obtained from the photographs and the frequencies calculated. These values of frequency, shown in Appendix I, were in agreement with those calculated by Cooper [1] for similar values of current, pressure, and tube diameter.

By dividing the period of rise by the total period of the striation, a ratio of rise time to total time was obtained. The ratio was found to vary approximately linearly with pressure. (See Appendix II) Because the data obtained for the experiment were taken primarily to determine a phase difference in lines of different excitation level, the first concern was that of obtaining stable striations. Due to the randomness of the frequency and current and limited ranges of pressure, it is difficult to say with any certainty that the ratio of rise time to total time does in fact vary linearly with the pressure. It is, however, clear that the ratio is dependent in some way upon the pressure.

4. Conclusions

4.1 Conclusions

There exists a phase difference at both the point of minimum and maximum intensity between lines of different excitation energy in the pressure range of 2mmHg to 7mmHg, the lower excitation leading the higher excitation line at minimum intensity and the higher excitation leading the lower excitation line at maximum intensity. These results do not necessarily conflict with those of Donahue and Dieke. [3] They compared lines of high excitation potentials with a line whose excitation energy was approximately the same as the metastable states of mercury. In order to do this in the case of argon, it would be necessary to work in the vacuum ultraviolet range of the spectrum.

The phase difference at maximum intensity is somewhat larger than those values previously reported by Pigg and Burton [9] for a 2cm diameter tube. They, however, used continuous sweep pictures to obtain their data. This method has been found by the author to give somewhat erroneous results because of the broadness of the striations in a continuous sweep picture.

Because of the phase difference, it appears that the wave causing the excitation (and thus the striation) must have a gradual frontal slope. This would cause the lower energy levels to be excited first. Now at peak intensity there will be transitions into the lower energy levels both from electrons cascading down from the upper levels as well as from excitation out of lower states. The combination of these two effects must cause the lower energy

lines to reach their maximum intensity at a later time than the lines of higher excitation. To understand what actually happens, one must have a knowledge of the excitation cross sections, the electron temperatures, the population of metastable atoms, and the lifetimes of the states.

There appears to be a relation between the wave shape of the striations and pressure. With only a limited amount of data over a very small pressure range, the ratio of rise time to the total period of the striation appears to increase linearly with pressure.

4.2 Recommendations for Further Work

More experimentation is required to measure the phase shifts between lines of higher and lower excitation at various pressures and tube sizes. It is suggested that some better method than the scanning of photographs be employed, as the time lag between collecting and correlating the data is extremely long. If one used conventional film, the negative in the form of film roll could be projected on a screen and the data removed directly.

The detailed shape and absolute intensity of the striation for lines of different excitation potentials appears to be a way of studying the form of the wave causing the striations. Experiments along these lines could be conducted using the equipment and techniques of this experiment.

4.3 Acknowledgements

The author wishes to express his appreciation to Professor R. L. Kelly for his advice and guidance, and to my wife for her patience and assistance in the scanning of the photographs.

I also express my appreciation to Mr. R. C. Moeller for rebuilding the vacuum system and for the technical assistance of Messieurs R. A. Garcia, K. Smith, and J. W. van Gastel, Jr.

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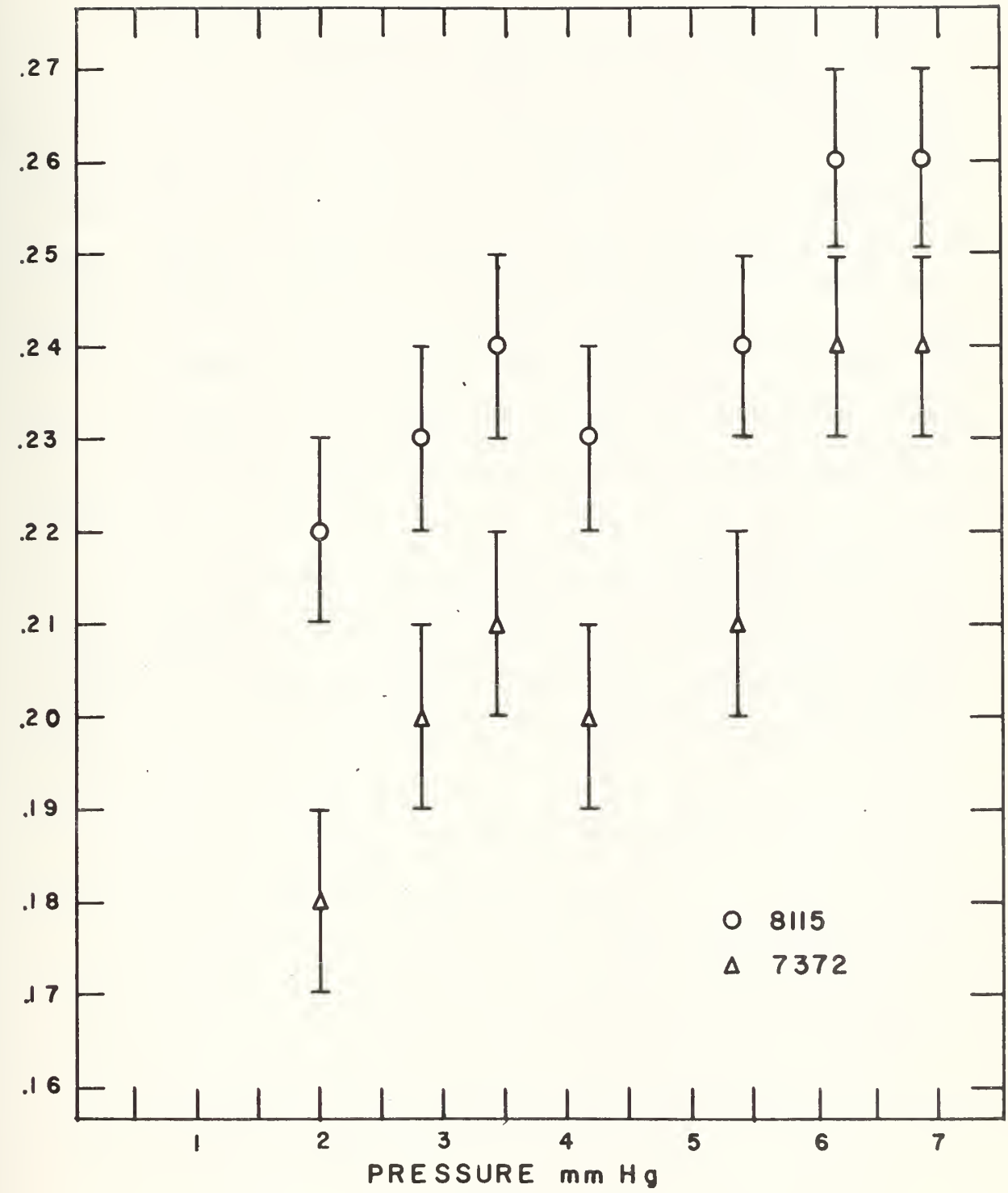
APPENDIX I

Phase Differences for the 8115A and 7372A Wavelengths

Pressure (mmHg)	Current (milli amps)	Phase Diff. at Minimum Intensity (microsec)	Phase Diff. at Maximum Intensity (microsec)	Frequency (cycles/sec)
2.02	220	$25^{+}_{-}8$	$35^{+}_{-}18$	$679^{+}_{-}9$
2.82	210	$29^{+}_{-}12$	$30^{+}_{-}19$	$591^{+}_{-}8$
3.43	250	$30^{+}_{-}12$	$34^{+}_{-}19$	$541^{+}_{-}4$
4.17	250	$30^{+}_{-}12$	$27^{+}_{-}18$	$579^{+}_{-}6$
4.91	210	$24^{+}_{-}13$	$34^{+}_{-}19$	-----
5.38	290	$23^{+}_{-}11$	$30^{+}_{-}17$	$600^{+}_{-}11$
6.18	300	$23^{+}_{-}9$	$27^{+}_{-}22$	$592^{+}_{-}8$
6.85	310	$25^{+}_{-}13$	$31^{+}_{-}14$	$601^{+}_{-}10$

Slit size 155 microns

APPENDIX II



RATIO OF RISE TIME τ TO PERIOD T VS PRESSURE FOR TWO DIFFERENT WAVELENGTHS

APPENDIX III

EXPERIMENTAL APPARATUS

1. Vacuum System

A schematic diagram of the vacuum and filling system is shown in Fig. 2, and a close-up view displaying the front panel and valve arrangement, filling system, and oil manometer in Fig. 4. The vacuum and filling system was modified during the course of this experiment from a glass to a metal system for increased pumping speed. The modified system consisted of the following equipment:

a. Type ED-35 Edwards Speedivac Fore Pump (1.25 c.f.m.).

b. Veeco 2" air cooled diffusion pump (85 l.p.s. at $5X10^{-5}$mmHg).

c. 2.5 liter copper trap attached to the top of the diffusion pump.

Pressure measurements in the system were made by two thermocouple gauges, one located above the main cut-off valve and the other between the diffusion pump and fore pump, and a Consolidated Vacuum Corp. type GIC-110 VGIA ionization gauge between the cold trap and main cut-off valve.

The system was capable of reducing the pressure to $5X10^{-7}$mmHg.

2. Discharge Tube and Gas Filling System

The discharge tube was made of pyrex glass tubing of 29.6mm O.D. and 25.6mm I.D. The overall length of the tube was 89.5cm with a distance of 71cm between electrodes. Each tube base was constructed in a similar manner with a tungsten filament surrounded by a cylindrical nickel electrode.

Prior to the installation of the tube on the vacuum system, it was placed on a portable vacuum system and baked (using heating tapes) for several hours. The electrodes were heated with an induction heater and the filaments were cleaned by running 6 amp currents through them. The process was repeated once the tube was installed on the permanent vacuum system. (This process was periodically repeated when the discharge became impure) The electrodes were then heated cherry red with the induction heater, and argon gas was passed into the tube allowing the electrodes to cool down in argon atmosphere.

Once the tube had gone through the above preparation, it was filled with argon and run several times until a pure discharge was obtained.

The argon gas was provided from two one-liter pyrex flasks of Linde high-purity gas which were sealed to the system. The gas filling was controlled by a small bulb between two stopcocks. The gas pressure was measured with a 100cm Octoil-S filled manometer (1cm oil=0.672mmHg).

Once the discharge tube was filled with gas, it could be isolated from the rest of the system by an Alpert ultra-high vacuum valve.

3. Power Supply

A direct current argon discharge was maintained by the following equipment: (See Figs. 3 & 5)

2. The discharge tube voltage was supplied by a Kepco Labs. model 770B power supply (0-600vDC). The tube current was measured with a built-in ammeter on the power supply (0-3 amp).

b. The auxiliary discharge at the anode end was supplied by a Kepco Labs. model 520R-B power supply (0-600v, 0-300ma).

c. The cathode was heated by a Kepco Labs. model KM 236-15A power supply (0-50v, 0-15amp).

4. Spectrograph and Measuring Instruments

A schematic of the physical arrangement is shown in Fig. 3. The light emitted to the spectrograph was constricted by a Gaertner adjustable slit (width set at 70 to 155 microns) placed in front of the tube. The slit was mounted on a stand which permitted both translation and rotation.

The photomultipliers used were an RCA 7102 (with a phototube made by Eldorado Electronics) for observing the 8115 and 7372A lines, and an RCA 1P21 (shielded with aluminum foil) for observing the 4158A line. Power was supplied to the phototubes by a Power Designs Pacific Inc., high voltage power supply (1-2000v, 0-15ma).

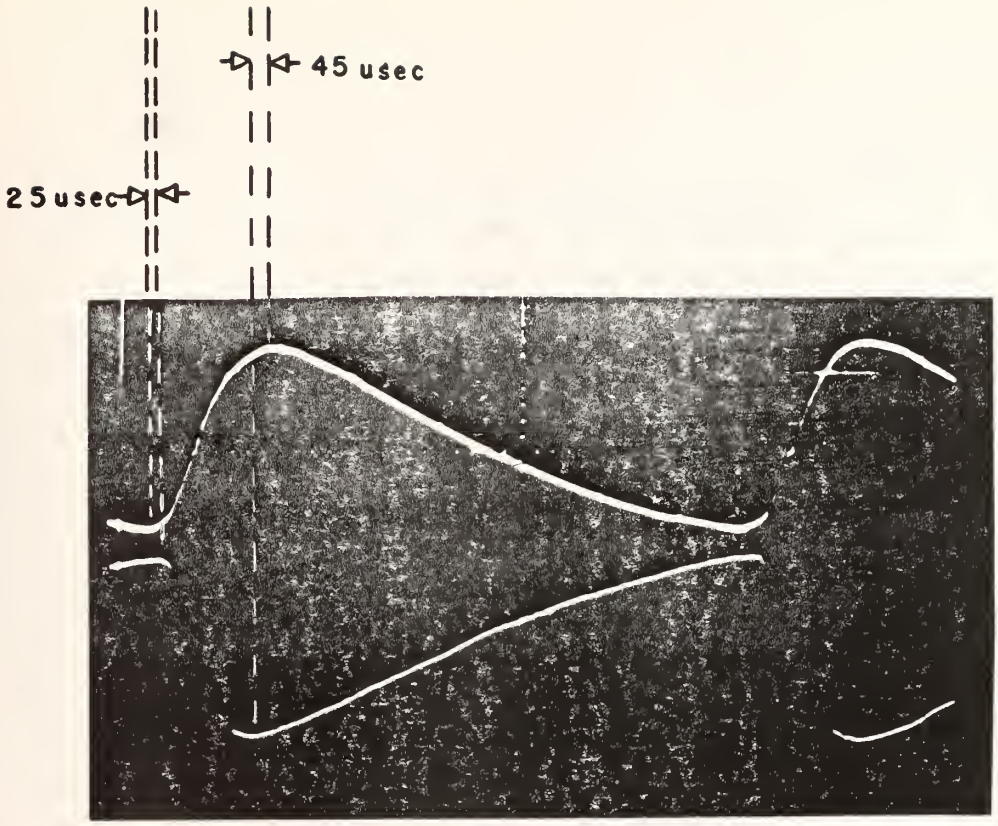
The signal outputs of the photomultipliers were hooked into two Tektronix type E, low level differential A. C. pre-amps (.05-10mv, 10 megohm input impedance shunted with a 50 50mm. capacitor, 6 microsec rise time, .06 cycles-60 kilocycles). The pre-amps were plugged into a Tektronix type 551 dual-trace oscilloscope. The striations were visually monitored, in addition to the scope presentation with a rotating mirror.

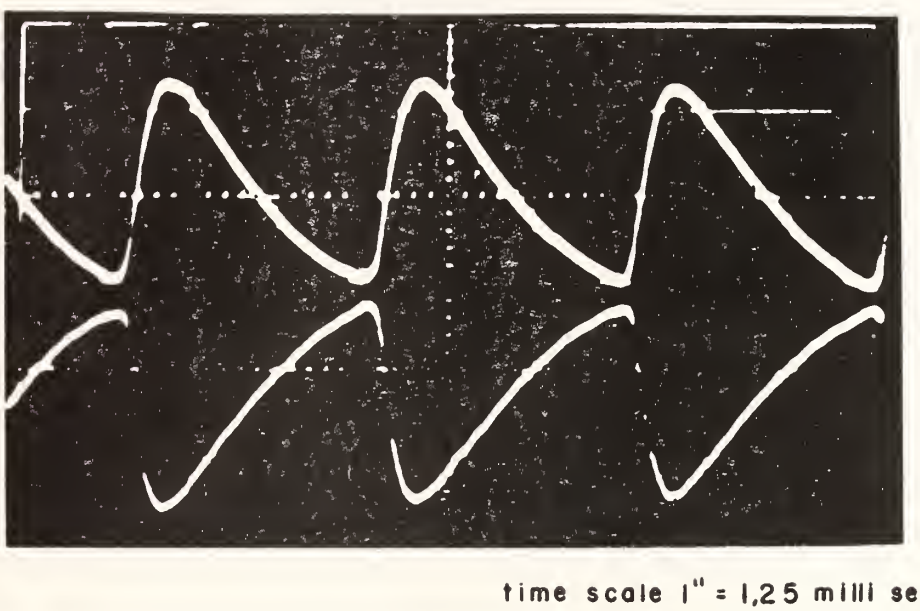
The spectrograph was a Baird-Atomic Plasma Spectrograph with a three-meter concave grating (on an Eagle mount), ruled with 15,000 lines per inch. The tube was mounted such that the discharge axis was perpendicular to the optical axis with the tube center aligned approximately with the entrance slit.

The spectrograph has a phototube rack designed for vertically mounted tubes which can be inserted into the spectrograph. Because it was necessary to use RCA 7102 tubes which must be mounted horizontally, the rack had to be modified. Consequently, the external movable feature of the rack could not be utilized, necessitating the adjustment of the tube placement by hand from within the spectrograph.

In order to ensure that only the desired spectral line fell on the phototube surface, a mask with small slits was mounted on the back of the phototube rack facing the grating. Additional slits were made for two intense visible spectral lines for purposes of aligning the strip with the grating. Once the mask was properly in place, these slits were sealed.

The sequence for the alignment of the spectral lines proceeded as follows, the signal being maximized in each step: (1) Phototubes were installed and adjusted. (2) The adjustable slit was placed in front of the tube, aligned, and slit width set. (3) The lens was adjusted. (4) Small adjustments were made in the grating setting by moving the gears manually in the rear of the spectrograph. This procedure was repeated when the equipment had remained idle for a few days.





PRESSURE 2 mm Hg

CURRENT .22 Amps

SLIT SIZE .155 mm

FIGURE 1 Oscilloscope Photographs of Striations

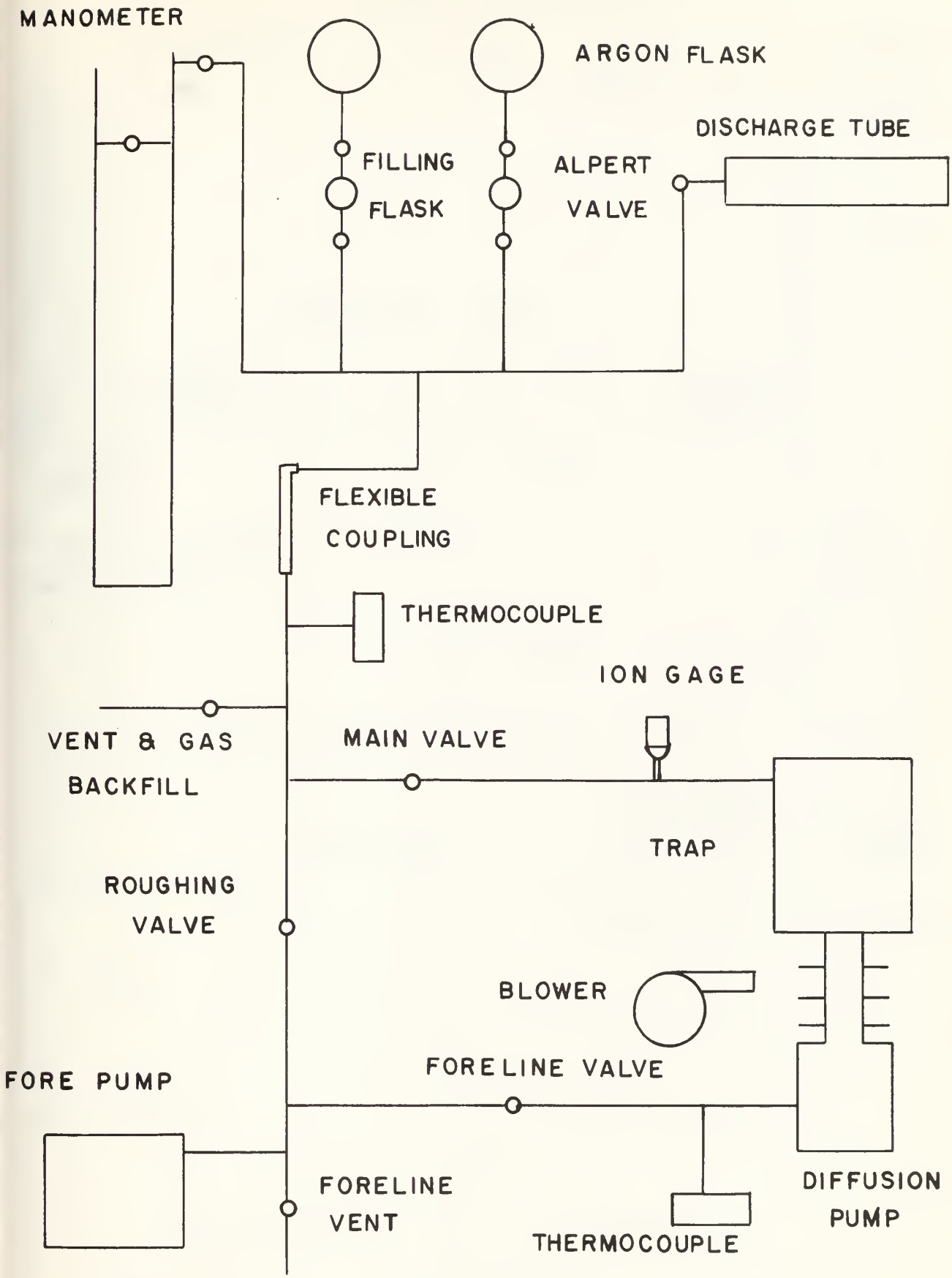


FIGURE 2 Schematic Diagram of Vacuum System and Filling System

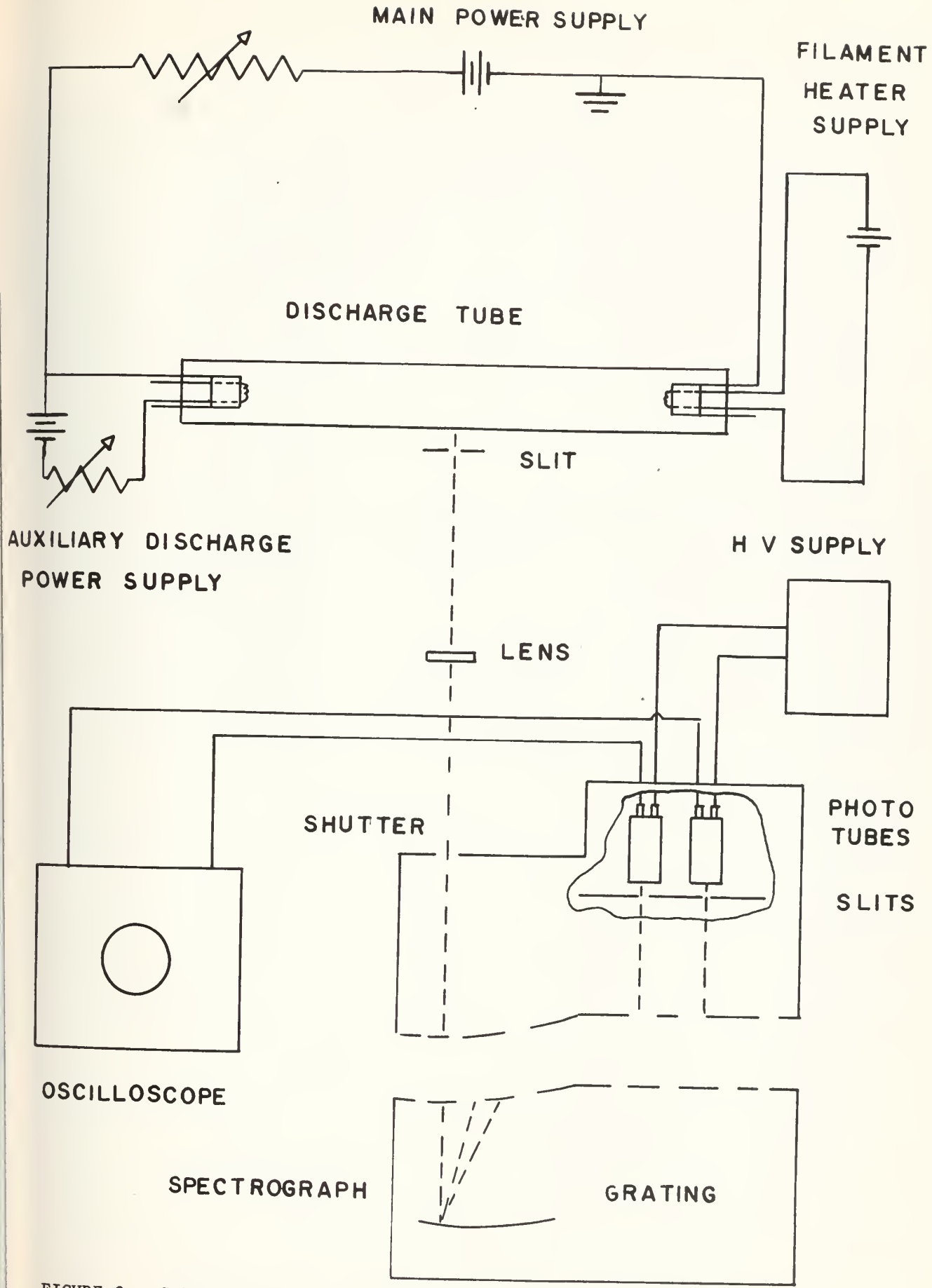


FIGURE 3 Schematic Diagram of Power Supply, Spectrograph and Measuring Instruments

Key to Apparatus in Figures 6 and 7

1. Fore Pump
2. Diffusion Pump
3. Liquid Air Trap
4. Front Panel and Valves
5. Manometer
6. Argon Flask
7. Rotating Mirror
8. Gaertner Slit and Stand
9. Discharge Tube
10. Meter for Ionization Gage and Thermocouple
11. H. V. Supply for Photomultipliers
12. Spectrograph
13. Power Supply for Discharge Tube
14. Power Supply for Heating Filament
15. Power Supply for Auxiliary Discharge

Figure 3 Overall View of Equipment

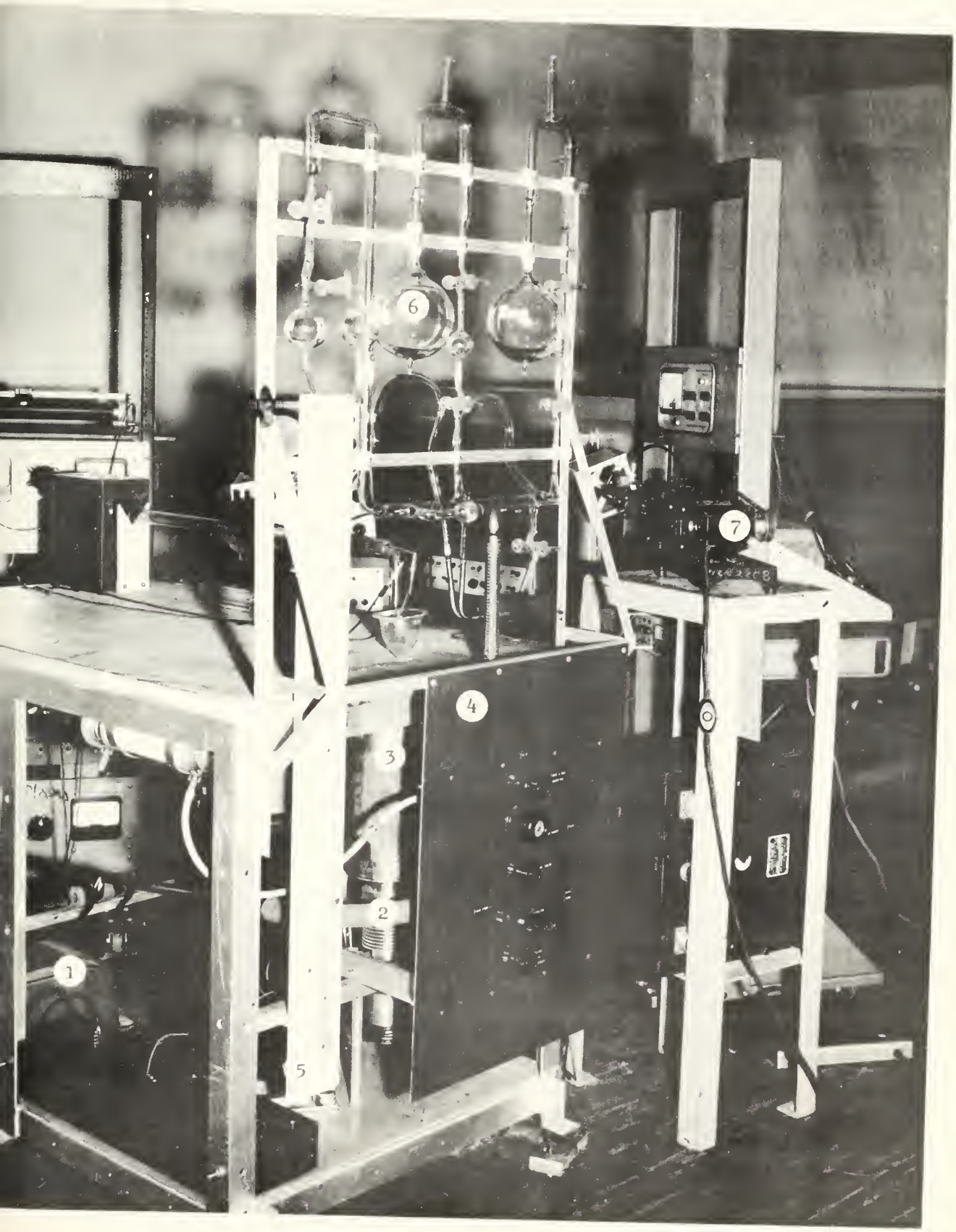


Figure 4 View of Vacuum System and Filling System